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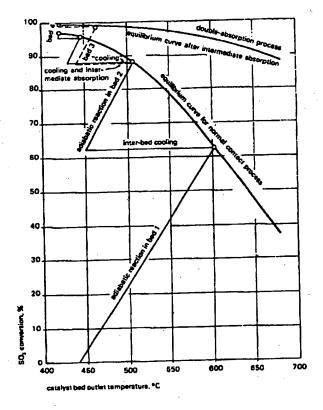
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(54) PROCEDE DE PRODUCTION D'ACIDE SULFURIQUE

(54) SULPHURIC ACID CONVERSION PROCESS



(57) A process for the production of sulphuric acid comprises providing a sulphuric acid precursor stream comprising oxygen and sulphur dioxide; feeding the sulphuric acid precursor stream to a sulphuric acid converter to produce a sulphuric acid stream and a gaseous stream containing unreacted sulphur dioxide; and, subjecting the gaseous stream to a regenerable sulphur dioxide recovery process to obtain a sulphur dioxide rich stream and a sulphur dioxide lean stream.

#### **Title: SULPHURIC ACID CONVERSION PROCESS**

#### FIELD OF THE INVENTION

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This invention relates to the production of sulphuric acid from sulphur dioxide.

#### **BACKGROUND OF THE INVENTION**

Much of the sulphuric acid which is produced today is obtained from the conversion of sulphur dioxide to sulphuric acid. Typically, the sulphur dioxide is first reacted in a sulphur dioxide converter with oxygen in the presence of a catalyst to produce sulphur trioxide. Subsequently, the sulphur dioxide is absorbed into a concentrated sulphuric acid solution (e.g. about 93 wt. % sulphuric acid) to produce sulphuric acid having a concentration of, for example, 96 - 98 wt. % acid.

The catalysts which are used for the conversion of sulphur dioxide to sulphur trioxide are often vanadium catalysts, such as vanadium pentoxide catalysts. These catalysts have a relatively narrow operating range. Generally, these catalysts have an ignition temperature of about 400 to about 420°C. Some high activity catalysts have been developed which have an ignition temperature of about 380°C. At temperatures above about 620 - 630°C, the catalyst becomes unstable and deactivates. Therefore, the sulphur dioxide must be at a temperature of at least about 380°C for the catalyst to become active and must not exceed a temperature of about 620 - 630°C in order to prevent degradation of the catalyst. Other catalyst types known in the art, such as platinum on a support carrier, may have different operating ranges and requirements.

The reaction of sulphur dioxide with oxygen to produce sulphur trioxide is an equilibrium reaction. It is also a highly exothermic reaction. The equilibrium of this reaction favours the production of sulphur trioxide at low temperatures. Further, as the reaction is exothermic, it is desirable to maintain the temperature of

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the sulphur dioxide which is introduced to the reactor as low as possible, while still achieving ignition, to prevent high temperature damage to the catalyst. Accordingly, the conversion efficiency of the reaction is furthered by maintaining a low concentration of sulphur dioxide in the feed stream. This necessitates the use of substantial quantities of a diluent gas and has several disadvantages. First, the increase in the amount of diluent gases causes a consequential increase in the amount of gas which must be processed through the sulphur dioxide converter thus necessitating a substantial increase in the size of the converter and the associated apparatus. The increase in the equipment size adds significantly to the cost of the sulphuric acid plant.

Secondly, sulphur dioxide is a pollutant which has been linked to acid rain and other environmental problems. Therefore, the permissible level of sulphur dioxide emissions have been reduced in recent years. In order to reduce the level of sulphur dioxide emissions, it is necessary to either obtain very high conversion in the acid plant or to treat any tail gases so as to reduce the concentration of sulphur dioxide. By increasing the amount of diluent gas which is used in the sulphur dioxide converter, there is a substantial increase in the amount of tail gas which must be treated and a consequential increase in the cost of operating a sulphuric acid plant. In particular, the plant equipment required to process the diluent gases must be increased due to the high amount of diluent gas which is required. The power consumed to transport these gases through the system, and to treat them, generally also significantly contributes to the cost of operating the sulphuric acid plant.

Various sulphur dioxide converters have been developed. In order to permit the autothermal operation of a converter, the feed gas to the sulphur dioxide converter should contain a minimum of about 5 - 6 wt. % sulphur dioxide for a single absorption process and 6 -

7 wt. % for a double absorption process. Further, currently utilized acid plant designs are limited to a maximum sulphur dioxide concentration of about 12 - 14 % by volume, in order to limit the catalyst temperature to less than its deactivation temperature.

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A further limit on the lower level of the sulphur dioxide concentration of the feed gas to the converter is the need to reduce the water/sulphur dioxide ratio in the feed gas to 1:1.07 or less in order to produce sulphuric acid of more than 93 wt. % concentration. If the acid has a concentration less than 93% (in the range of 78 - 93 wt. %), it becomes highly corrosive and impractical for a commercial embodiment. The water content of the feed gas is adjusted by cooling to the required temperature to condense out excess water. With dilute sulphur dioxide feed gases, this may require chillers which have a high capital cost and consume substantial quantities of power during the operation.

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In order to meet these competing requirements, sulphuric acid plants typically utilize sulphur dioxide converters which contain typically 3 - 6 catalyst beds, with cooling of the process gas between the catalyst beds. Most modern plants are of the double absorption type, which have an intermediate sulphur trioxide absorption tower that shifts the chemical equilibrium towards higher conversion. With these multiple pass units, it is possible to obtain tail gases which meet current pollution guidelines. With modern techniques, a conversion rate of about 80 volume per cent of the sulphur dioxide may be obtained in the first pass through a catalyst bed. Thus, a majority of the capital investment is directed towards the remaining catalyst beds (i.e. the second through fifth catalyst beds) to convert the remaining 20 vol. % of the sulphur dioxide to meet the EPA standard of 99.75 vol. % conversion of the sulphur dioxide to sulphur trioxide. An example of the complexity of equipment which is required by this approach is shown in Figures 1 and 2.

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Figure 1 is a comparison of the reactive profiles and sulphur dioxide conversion for a 4-bed normal contact (single-absorption) process and a (2+2) double-absorption process wherein the feed gas has a concentration of 8.5 volume percent sulphur dioxide. As can be seen from Figure 1, well over half of the sulphur dioxide is converted in the first catalyst bed. In the single absorption process, the exit gases from each catalyst bed are cooled before they are introduced into the next catalyst bed thus preventing excessive heating of the catalyst and enabling the catalyst to effect further conversion. As shown in dotted outline, in the double absorption process, the absorption of the sulphur dioxide after the second pass permits substantially greater conversion on the third catalyst pass due to the removal of the sulphur trioxide product of reaction thereby minimizing the need for cooling between the third and fourth catalyst beds. A typical double absorption process acid plant is shown in Figure 2. Sulphur and oxygen are fed to a sulphur furnace 10. To this end, air is fed by a stream 14 through air filter 16 into drying tower 18. The treated air is fed via blower 20 to furnace 10 where it is combusted with sulphur feed stream 12 to produce sulphur dioxide stream 22. The sulphur dioxide stream may be fed directly, or optionally through waste heat boiler 24, to sulphur dioxide converter 26. Sulphur dioxide converter 26 has four catalyst beds 28, 30, 32 and 34. The product stream from the first catalyst bed 28 is fed to heat exchanger 36 to cool the product gases before the product gases are introduced to the next catalyst bed 30. The product gases from catalyst bed 30 are cooled in heat exchanger 37 and then introduced to catalyst bed 32. The product gases from the third catalyst bed 32 are fed to intermediate heat exchangers 38 and intermediate absorber 40 before being introduced into the fourth catalyst bed 34. The product gases from the final catalyst bed 34 are fed to a heat exchanger 42 and then to final absorber 44. In order to prevent the introduction of excessive acid mist into the fourth catalyst bed,

intermediate absorber is provided with a mist eliminator 46. Acid mist is eliminated from tail gas stream 48 by candle filters 50. Each absorber 40 and 44 produces an acid stream 52 which is fed to acid tank 54.

In order to utilize the heat produced during the sulphur dioxide conversion process, boiler feed water 56 is fed to steam drums 58 and heat exchangers 36 and 42 to produce steam 60.

The use of multiple catalyst beds, heat exchangers and absorbers adds substantially to the capital cost of the sulphuric acid plant as well as the operating cost of such a plant.

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#### SUMMARY OF THE PRESENT INVENTION

In accordance with the instant invention there is provided a process for the production of sulphuric acid comprising the steps of:

- (a) providing a sulphuric acid precursor stream comprising oxygen and sulphur dioxide;
- (b) feeding the sulphuric acid precursor stream to a sulphuric acid converter to produce a sulphuric acid stream and a gaseous stream containing unreacted sulphur dioxide; and,
- (c) subjecting the gaseous stream to a regenerable sulphur dioxide recovery process to obtain a sulphur dioxide rich stream and a sulphur dioxide lean stream.

In one embodiment, at least a portion of the sulphur dioxide rich stream is recycled and is fed to the sulphur dioxide converter. Preferably, all of the sulphur dioxide rich stream is recycled and fed to the sulphuric acid converter.

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The sulphuric acid converter preferably utilizes two catalyst beds and, more preferably, a single catalyst bed. The gaseous stream exiting the catalyst beds is fed to a sulphur trioxide contactor (eg. an absorption tower) where the sulphur trioxide is absorbed into a concentrated acid stream (preferably at least about 93 wt. % sulphuric acid and, more preferably, about 93 wt. % sulphuric acid). The gases

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leaving the absorption tower are preferably fed to the regenerable sulphur dioxide recovery process. Accordingly, one advantage of the instant process is that it is not necessary to subject the tail gases to a candle filter to eliminate acid mist from being released to the atmosphere.

Further, and more importantly, by eliminating the need to treat the emissions from the sulphur trioxide absorber as tail gases which will be released to the atmosphere, it is no longer needed to maximize the conversion of the sulphur dioxide to sulphur trioxide during a single pass through a catalyst bed or a series of passes through multiple catalyst beds. In particular, the sulphur dioxide converter may comprise only one or two catalyst beds. These are the catalyst beds wherein the majority of the sulphur dioxide conversion to sulphur trioxide occurs. The product gases, which still contain a significant amount of sulphur dioxide, after being passed through the sulphur trioxide absorber, may be recycled to the sulphuric converter. A portion of these gases may be fed to the regenerable sulphur dioxide recovery process to increase the concentration of the sulphur dioxide in the feed stream to the sulphuric acid converter. More preferably all of these gases are treated by the regenerable sulphur dioxide recovery process. It will be appreciated that by eliminating the need for a substantial number of catalyst beds (e.g. up to six), the sulphuric acid plant is substantially simplified. For example, fewer heat exchangers, pumps, blowers and conduits are required. Further, the need for multiple absorption towers may also be avoided.

As the unreacted sulphur dioxide is recycled back to the sulphur dioxide converter, the sulphur dioxide converter may be run using the highest possible SO<sub>2</sub> concentration in the feed, as permitted by the required oxygen to sulphur dioxide ratio. In particular, the feed gas which is fed to the sulphur dioxide converter may contain more than about 12 volume percent sulphur dioxide, preferably more than

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14 volume percent sulphur dioxide, more preferably more than about 16 volume percent sulphur dioxide, and, most preferably more than about 18 volume percent sulphur dioxide. This minimizes the volume of feed gas which is fed to the converter and therefore reduces the size and cost of the sulphur dioxide converter and the absorption tower.

In accordance to another embodiment of the instant invention, the process for the production of sulphuric acid comprises the steps of:

- (a) providing a first feed stream comprising sulphur dioxide:
- (b) subjecting at least a portion of the first feed stream to a regenerable sulphur dioxide recovery process to obtain a first sulphur dioxide rich stream and a first sulphur dioxide lean stream;
- (c) feeding at least a portion of the first sulphur dioxide rich stream and a second feed stream comprising oxygen to a sulphur dioxide converter to produce a product stream containing sulphur trioxide; and,
- (d) contacting the product stream with an aqueous solution to produce a sulphuric acid stream and a gaseous stream containing unreacted sulphur dioxide and inert gases.

The gaseous stream may be subjected to the regenerable sulphur dioxide recovery process to obtain a second sulphur dioxide rich stream and a second sulphur dioxide lean stream. At least a portion of the second sulphur dioxide rich stream, and preferably all of it, is recycled and fed to the sulphur dioxide converter.

In one embodiment, a portion of the feed stream containing sulphur dioxide is fed directly to the sulphur dioxide converter. In another embodiment, all of the feed gas stream is preferably subjected to the regenerable sulphur dioxide recovery process.

The sulphur dioxide lean streams preferably contain less

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than about 300 ppm sulphur dioxide, more preferably less than about 150 ppm sulphur dioxide and, most preferably less than about 100 ppm sulphur dioxide.

In some operations, the concentration of sulphur dioxide in the sulphur dioxide feed stream may vary. If the concentration of the feed gas which is fed to the sulphur dioxide converter varies too greatly, the sulphur dioxide converter will not operate efficiently. Therefore, in accordance with another embodiment of this invention, there is provided a method of operating a sulphuric acid plant comprising:

- (a) providing a feed stream comprising sulphur dioxide;
- (b) monitoring the sulphur dioxide concentration of the feed stream;
- (c) if the sulphur dioxide concentration of the feed stream is below the level required to achieve autothermal operation of a sulphur dioxide converter, subjecting at least a portion of the feed stream to a regenerable sulphur dioxide recovery process to obtain a first sulphur dioxide rich stream having a sulphur dioxide concentration sufficient to permit autothermal operation of the sulphur dioxide converter and a first sulphur dioxide lean stream;
- (d) feeding a stream selected from the group consisting of at least a portion of the feed stream, at least a portion of the first sulphur dioxide rich stream and mixtures thereof to the sulphur dioxide converter to produce a product stream containing sulphur trioxide and unreacted sulphur dioxide; and,
- (e) contacting the product stream with an aqueous solution to produce a sulphuric acid stream and a gaseous stream containing unreacted sulphur dioxide.

#### 30 DESCRIPTION OF THE DRAWING FIGURES

These and other advantages of the instant invention will be

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more fully and completely understood in connection with the following description of the drawings in which:

Figure 1 is a comparison of the reaction profiles and sulphur dioxide conversion for a prior art four-bed normal contact (single absorption) process and four-bed (2+2) double-absorption

Figure 2 is a prior art plant schematic for a sulphur-burning process; double-absorption sulphuric acid process;

Figure 3 is a schematic of a sulphuric acid plant according to a preferred embodiment of this invention; 10

Figure 4 is a schematic for a sulphur dioxide plant according to the instant invention wherein the sulphur dioxide converter comprises a two bed converter with a split feed;

Figure 5 is an alternate embodiment showing a sulphur dioxide converter comprising a single pass wet catalyst bed with

Figure 6 is a further alternate embodiment showing a recycle; sulphur dioxide converter utilizing a single pass, dry catalyst bed with

Figure 7 is a further alternate embodiment showing a recycle; sulphur dioxide converter having two catalyst bed sulphur dioxide

Figure 8 is a schematic diagram of a regenerable sulphur converter; and, dioxide process which may be used in the instant invention.

## DESCRIPTION OF PREFERRED EMBODIMENT

Referring to Figure 3, a process according to the instant invention comprises a regenerable sulphur dioxide recovery unit 100 and an acid conversion unit 102. Feed gas 104 containing sulphur dioxide is fed to the process to produce concentrated acid stream 106 and inert or sulphur dioxide lean stream 108. Impurities are removed from the system via stream 98.

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Sulphur dioxide plants are typically used to convert sulphur dioxide to a commercial product, namely sulphuric acid. Therefore, the sulphur dioxide in feed stream 104 may be from any source known in the industry. For example, feed stream 104 may be the combustion stream from a sulphur burner wherein sulphur is burned in the presence of oxygen to produce sulphur dioxide. Alternately, stream 104 may comprise one or more of flue gases from the combustion of sulphur bearing fuels, metallurgical gas from the roasting of various sulphur containing ores, recovery boiler flue gas fired with spent sulphite cooking liquors, the tail gas from the incineration of sulphur containing non-condensible gas produced by kraft pulping, incinerator gas from the burning of spent, contaminated sulphuric acid or the vent and digester blow gases of a sulphite pulp mill.

The sulphur dioxide content of these kinds of gases can vary from less than a few thousand ppm of sulphur dioxide to almost 100 vol. % sulphur dioxide. Water and various impurities may also be present such as sulphur trioxide, sulphuric acid mist, nitrogen oxides, ash, volatile metals or metal oxides, carbon, carbon monoxide, organic compounds and the like. These gases may be at various temperatures and pressures although they are typically close to atmospheric pressure.

Feed stream 104 may be fed to the regenerable sulphur dioxide recovery unit 100. Alternately, if feed stream 104 contains a sufficient level of sulphur dioxide to permit the autothermal operation of the catalyst beds of conversion unit 102 (e.g. greater than about 5 - 7 % sulphur dioxide), and it does not contain any particulate or other matter which is undesirable in the acid conversion unit 102 in order to prevent damage to the catalyst in the catalyst bed, such as fluorine, chlorine or arsenic, some or all of feed stream 104 may be fed via by-pass stream 110 directly to conversion unit 102.

In accordance with the instant invention, the feed gas which is fed to conversion unit 102 may contain more than 12 volume percent SO<sub>2</sub>, preferably more than 14 volume percent SO<sub>2</sub>, more preferably more than 16 volume percent SO<sub>2</sub> and, most preferably, more than 18 volume percent SO<sub>2</sub>. In order to increase the concentration of sulphur dioxide in the feed gas to conversion unit 102, some or all of stream 104 may be fed to recovery unit 100.

It will be appreciated that the process gas streams of the instant invention are preferably also treated as is known in the art to maintain a sufficient ratio of oxygen to sulphur dioxide as well as the ratio of water to sulphur dioxide to result in the production of a solution having 93 wt. % or more sulphuric acid.

Sulphur dioxide recovery unit 100 may utilize a number of processes which are known in the art. For example, the regenerable process may comprise the Selexol™ process as described in Hydrocarbon Processing, April 1996, p. 133; the Solinox™ process as described in Heisel M.P. and Belloni A.E., Gas Separation and Purification, vol. 5: p. 111 ff (June 1991); the Wellman-Lord process as described in Sulphur, Sulphur Dioxide and Sulphuric Acid, English Edition prepared by A. I. More, p. 235, The British Sulphur Corporation (1984); the regenerable amine-based sulphur dioxide scrubbing process of Dow Chemical as described in United States Patent No. 5,236,678; and, the Cansolv™ System sulphur dioxide scrubbing process as described in United States Patent Numbers 5,019,361 and 5,262,139. Preferably, recovery unit 100 utilizes the Cansolv™ System Process.

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Figure 8 shows a schematic diagram of the Cansolv<sup>TM</sup> process. Pursuant to this preferred embodiment, recovery unit 100 utilizes an absorber 112 and a regenerator 114. Feed gas 104 is fed to pre-scrubber 116 where it is contacted with recirculated water from a spray tower 118. This cools and saturates feed gas 104 and results in the

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removal of a large fraction of the particulate matter in feed gas 104, depending upon their size. Strong acids such as sulphuric and hydrochloric acid may also be scrubbed. The pre-scrubber can also be used as a direct cooler by adding a heat exchanger to the water circulation loop. The level of dissolved acids in the pre-scrubber is controlled by blow-down, neutralizing and discharge into a waste water treatment system. Suspended solids my be controlled by settling.

The pre-scrubbed feed gas is then fed to absorber 120. Absorber 120 is preferably a mass transfer device for contacting an absorbent with the treated feed gas. As shown in Figure 8, a counter-current contacting system may be utilized. Any conventional absorber may be used, such as a packed or trayed tower. The scrubbed feed gas which has the sulphur dioxide removed therefrom may be fed via stream 128 to, for example, a stack for discharge to the atmosphere. The exact process to which stream 128 will be subjected will vary depending upon the content of the stream. Preferably, stream 128 contains less than 300 ppm sulphur dioxide, more preferably less than 150 ppm sulphur dioxide and, most preferably less than 100 ppm sulphur dioxide.

According to the Cansolv™ process, the feed gas stream is contacted with a water-soluble half salt of a diamine at a temperature of from about 10° to about 60°C to absorb sulphur dioxide from the gas stream in amounts of at least about 100 grams of sulphur dioxide per kilogram of absorbing medium. The diamine in free base form preferably has a molecular weight of less than 300 and, in half salt form, has a pKa value for the free nitrogen atom of about 5 to about 7.3.

The rich  $SO_2$  laden absorbent stream 130 from absorber 120 is pumped to regenerator 114 via a rich/lean heat exchanger 122. Regenerator 114 may be a trayed tower with a steam heated re-boiler 124. As the absorbent flows down regenerator 114, the sulphur dioxide

is stripped from the liquid and carried overhead into a reflux condenser 132 where most of the steam condenses and is returned to the top of the regenerator as reflux stream 134. The gaseous sulphur dioxide may be stripped from the spent absorbing medium at a temperature of about 50 to about 110°C. The gaseous, water saturated sulphur dioxide leaves the regenerator as stream 136. The lean absorbent leaves re-boiler 124 via stream 138 and is pumped back to absorber 120 via heat exchanger 122 and a surge tank 126.

A slip stream 142 of the lean absorbent is preferably fed to absorbent purification unit 140. In unit 140, heat stable anions, such as sulphate, are replaced by regenerable sulphite anions sourced from reflux 134.

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Sulphur dioxide feed stream 144, oxygen feed stream 146 and water feed stream 148 are fed to acid conversion unit 102. In acid conversion unit 102, the sulphur dioxide is converted to sulphur trioxide and then absorbed in concentrated acid (e.g. preferably greater than about 93 wt. % sulphuric acid) to form a concentrated sulphuric acid stream (e.g. preferably 96 to 98 wt. % sulphuric acid or more). While this process may be accomplished by any means known in the art, preferably, acid conversion unit 102 utilizes two and preferably one catalyst bed.

The tail gases from acid conversion unit 102 are preferably recycled. As shown in Figure 3, some or all of tail gas stream 150 may be fed through recovery unit 100 to provide a concentrated sulphur dioxide stream. Alternately, if tail gas stream 150 contains few inerts such as would be the case if feed stream 146 comprised essentially pure oxygen, a portion or all of recycle stream 150 may be recycled and combined with stream 144 which is fed to acid conversion unit 102.

It will be appreciated by those skilled in the art that, depending upon the capacity of acid conversion unit 102, some or all of sulphur dioxide stream 144 and/or recycle stream 150 may be fed to

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a storage device (not shown) for storage for later use or it may be fed to another part of the plant where there may be a requirement for a sulphur dioxide stream.

In the embodiment shown in Figure 4, acid conversion unit 102 comprises a sulphur dioxide converter 152 having a first catalyst bed 156 and a second catalyst bed 154. All of the feed air stream 146 and part of the sulphur dioxide feed stream 144 are fed via split stream 158 to heat exchanger 160. The heated feed gases 162 are then fed via stream 162 to first catalyst bed 156. Product gases from the first catalyst bed are fed via stream 164 to heat exchanger 160 where they are used to heat the incoming feed gases from split stream 158. The cooled product gases (stream 166), which typically contain unreacted sulphur dioxide, are combined with the remaining unreacted sulphur dioxide feed in stream 168 and fed to second catalyst bed 154 to produce product gases 170.

The use of a split feed is advantageous if a feed gas containing a concentration of 14 vol. % sulphur dioxide or greater is available. In such a case, the use of a split stream feed, whereby some of the sulphur dioxide is fed to a second catalyst bed, is advantageous to maintain the exit temperature of the product gases (streams 164 and 170) below the degradation temperature of the catalyst. To this end, part of the sulphur dioxide feed gas (stream 158) is preferably passed through heat exchanger 160 to raise the temperature of heated feed gas stream 162 to the ignition temperature of the catalyst so that the converter will operate autothermaly. The sulphur dioxide concentration of heated feed gas stream 162 is preferably the maximum concentration which will maintain product gases from the first catalyst bed (stream 164) below the degradation temperature of the catalyst. The heated gases are cooled by passing them through heat exchanger 160 where they heat incoming feed stream 158. The cooled product gases (stream 166) may then be combined with the remaining sulphur

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dioxide which has not yet been reacted (stream 168) which then pass through the second catalyst bed 154.

Product stream 170 contains sulphur trioxide, unreacted sulphur dioxide as well as inert gases (e.g. nitrogen) if oxygen feed stream 146 comprises air. It will be appreciated that if oxygen enriched air is used as oxygen feed stream 146 or, alternately if oxygen feed stream 146 is pure oxygen, then the amount of nitrogen in feed stream 170 will be substantially less or may in fact be negligible.

Product gas stream 170 is at an elevated temperature (e.g. about 600°C). Accordingly, it preferably cooled before being subjected to a sulphur trioxide absorption process. Accordingly, as shown in Figure 4, product gas stream 170 may be fed to a waste heat boiler 172 where it is used to heat boiler water 174 to produce steam 176. The cooled product gases exit waste heat boiler 172 via stream 178 and are fed, such as by fan 180, to sulphur dioxide absorption tower 182. The sulphur dioxide is absorbed into a aqueous solution, preferably at least 93 wt. % sulphuric acid which is introduced into absorption tower 182 via stream 148. Typically, the cooled product gases pass counter-current to the aqueous stream 148 to produce a concentrated acid stream 184 which is fed to acid tank 186. The concentrated acid stream may comprise, for example, 96 to 98 wt. % sulphuric acid. The actual concentration of acid stream 184 may vary depending upon the desired concentration of acid which is to be produced by the plant.

The acid in tank 186 may be mixed with the incoming concentrated acid 184 by means of motor 188 which may continuously stir the liquid in acid tank 186. Make-up water 190 may be added to acid tank 186 to alter the concentration of the acid.

The concentrated acid in acid tank 186 exits acid tank 186 via stream 192. Stream 192 may be cooled by passing it through heat exchanger 194 subsequent to which some of the acid is drawn from the process to obtain product acid stream 106. The remainder is fed to the

top of absorption column 182 via stream 148. Alternately, some or all of stream 192 may be fed via by-pass stream 196 to absorption column 182 if shut off valve 198 is open.

Tail gas stream 150 exits absorption column 182. A portion of tail gas stream 150 may be recycled through converter 152 to convert further sulphur dioxide present in tail gas stream 150 to sulphur trioxide. Accordingly, tail gas stream 150 may be combined with sulphur dioxide feed stream 144 and fed directly back to converter 152 (as shown by the broken line in Figure 4). Alternately, a portion of tail gas stream 150 may be fed to the regenerable sulphur dioxide recovery unit. The recovered sulphur dioxide may then form part of feed stream 144. In a preferred embodiment, all of tail gas 150 is fed to the regenerable sulphur dioxide recovery unit 100 wherein a concentrated sulphur dioxide stream may be obtained (as shown in Figure 3).

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If oxygen feed stream 146 comprises pure oxygen, then tail gas stream 150 will substantially comprise sulphur dioxide and oxygen. Therefore, in such an embodiment, it is preferred to recycle tail gas stream 150 directly to converter 152 without feeding tail gas stream 150 to absorber 112. Accordingly, it will be understood that a further advantage of the instant invention is that oxygen may be economically used as the oxygen source for converter 152 as the oxygen, as well as the sulphur dioxide would essentially be recycled to extinction (i.e. until they are effectively fully consumed). It will be appreciated that a bleed stream or the like may be required to remove inert compounds and other by-products which will build up in the system.

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It will also be appreciated that if the oxygen stream 146 comprises air, that tail gas stream 150 is preferably fed to absorber 112 so as to remove the nitrogen from the system.

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Figure 5 shows an alternate process schematic wherein converter 152 comprises a single catalyst bed 154. Figure 5 shows a schematic for a wet catalysis process. The sulphur dioxide feed gas

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stream 144 and the oxygen feed gas stream 146 are combined to produce precursor stream 200 which is fed, for example, by blower 202 into catalyst bed 154 to produce product gas stream 204 which contains unreacted sulphur dioxide, oxygen, sulphur trioxide, other by-product gases as well as nitrogen if air is used in oxygen feed stream 146.

A portion of product gas stream 204 is recycled via stream 206 and mixed with mixed stream 200 prior to the introduction of stream 200 into converter 152. By recycling stream 206 and mixing it with unreacted mixed stream 200, the effective concentration of sulphur dioxide in the feed gases to converter 152 is reduced. Accordingly, recycle stream 206 effectively dilutes the concentration of sulphur dioxide in the feed gases thereby serving to moderate the exit temperature of product gases 204 to prevent deactivation of the catalyst from overheating. Further, by directly mixing recycle stream 206 with unreacted mixed stream 200, the unreacted gases may be raised to the ignition temperature of the catalyst.

Figure 6 shows a schematic of an alternate embodiment wherein converter 152 has a single catalyst bed 154 and the feed gases are dried prior to entering converter 152. Accordingly, feed gas streams 144 and 146 may be fed to dryer 208. The feed gases may be dried, for example, by passing them through dryer 208.

A further alternate embodiment is shown in Figure 7. Pursuant to this embodiment, converter 152 comprises a first catalyst bed 154 and a second catalyst bed 156. By-pass stream 210 is withdrawn from mixed stream 200 so that only part of the sulphur dioxide and oxygen containing stream is fed to first catalyst bed 154 to produce product gas stream 204. A portion of product gas stream 204 is recycled via stream 206 and mixed with mixed stream 200 prior to the entry of the unreacted gases into first catalyst bed 154 as discussed above with respect to Figure 5. The remaining product gases and the by-pass gases 210 are mixed together and fed to second catalyst bed 156 to produce

product gas stream 170. Accordingly, this embodiment permits a portion of the product gases from the first catalyst bed to be recycled to bring the portion of the mixed stream 200 to the ignition temperature for reaction to occur over the first catalyst bed. Further, the exit temperature of product gases 204 are also moderated by feeding a portion of the unreacted sulphur dioxide directly to the second catalyst bed 156 via by-pass stream 210 in a similar manner as discussed with respect to Figure 4.

It will be appreciated by those skilled in the art that other configurations for converter 152 as well as for absorption column 182 and acid tank 186 may be utilized. For example, converter 152 could comprise a tubular isothermal reactor or a fluidized bed isothermal reactor. These two reactor types can convert feed streams higher in sulphur dioxide and oxygen without catalyst deactivation since a part of the reaction isotherm is removed to maintain the catalyst temperature below its deactivation temperature.

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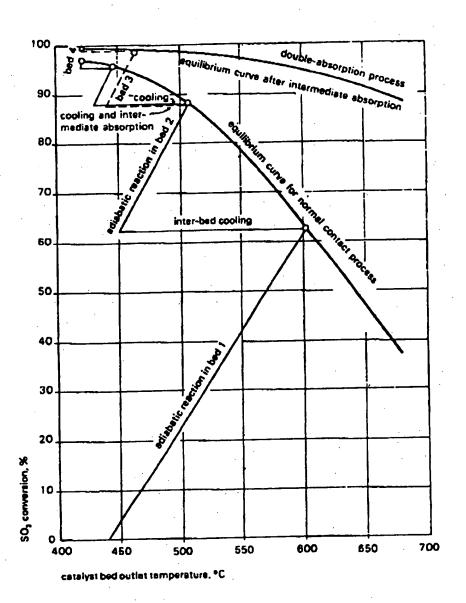
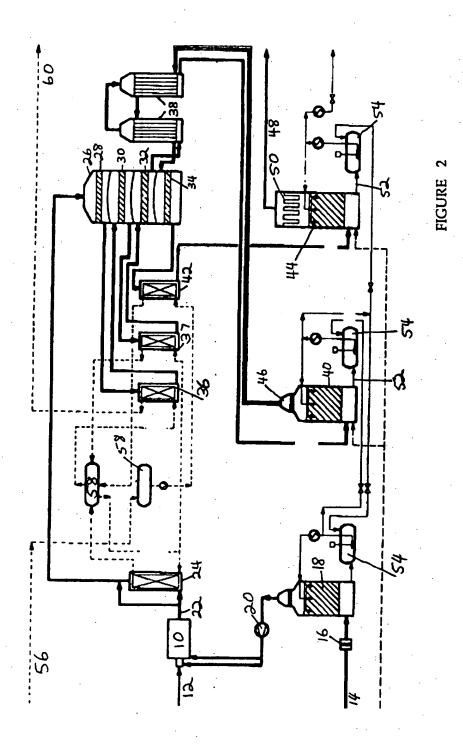
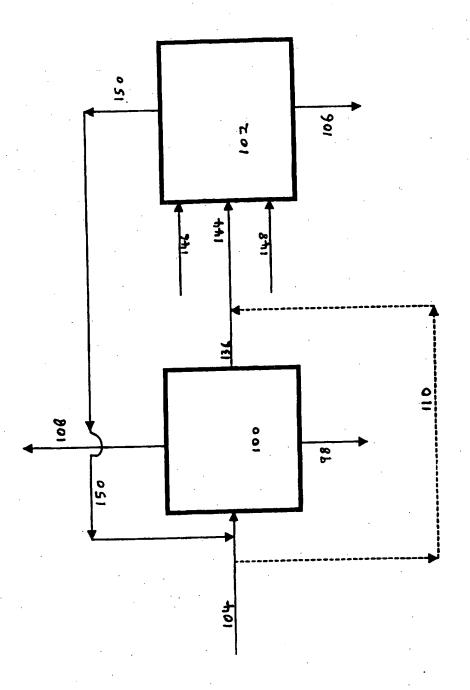
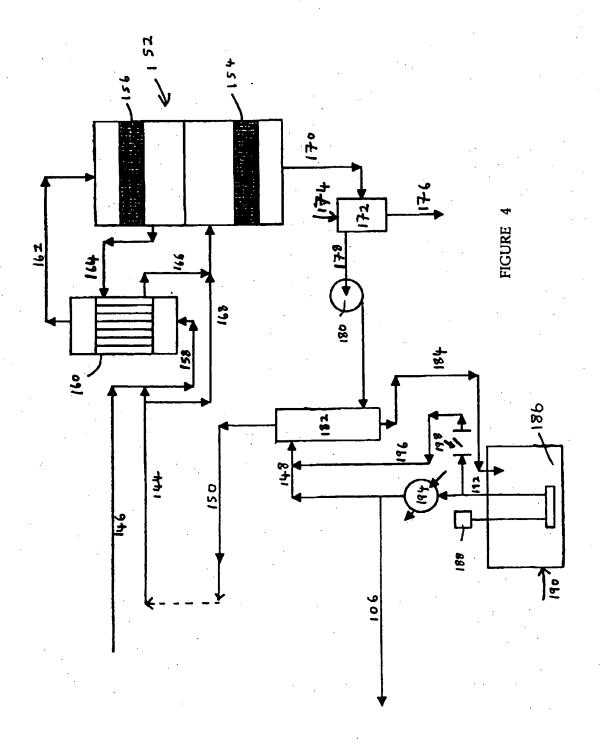
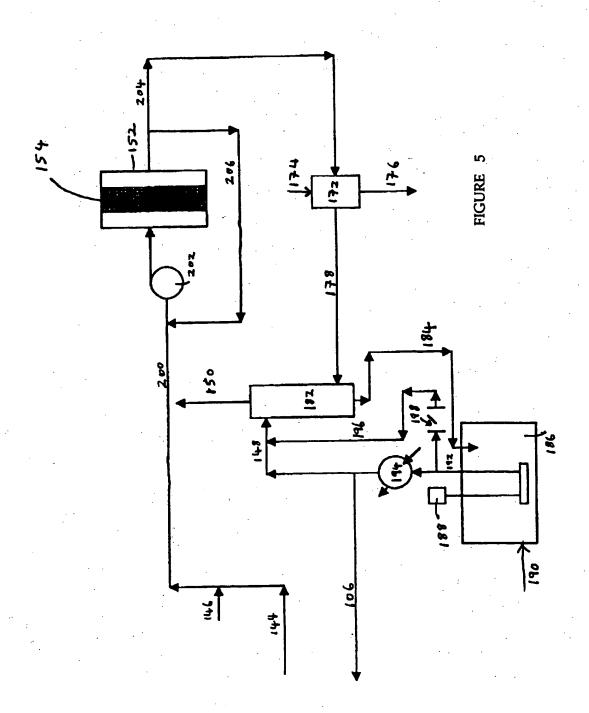


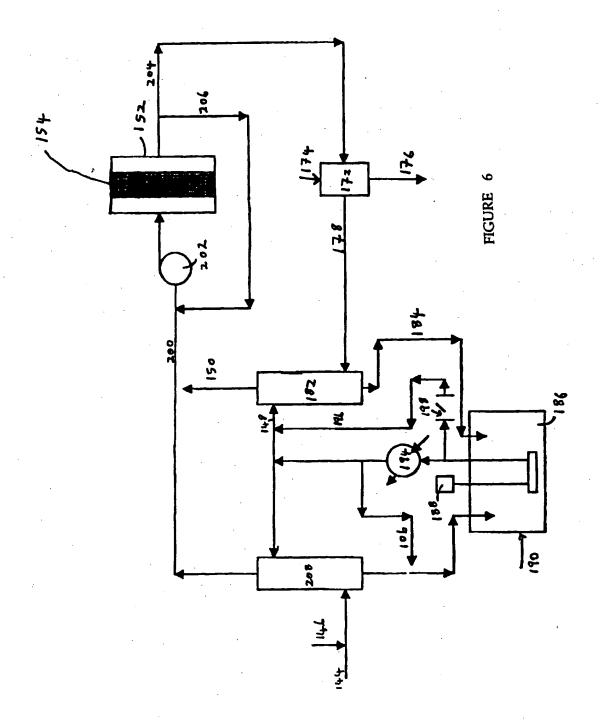
FIGURE 1











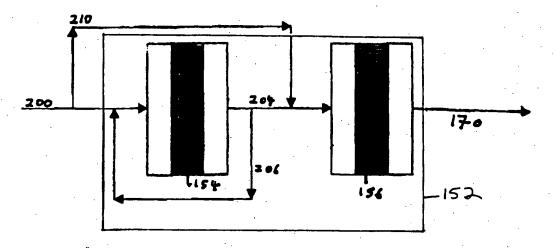
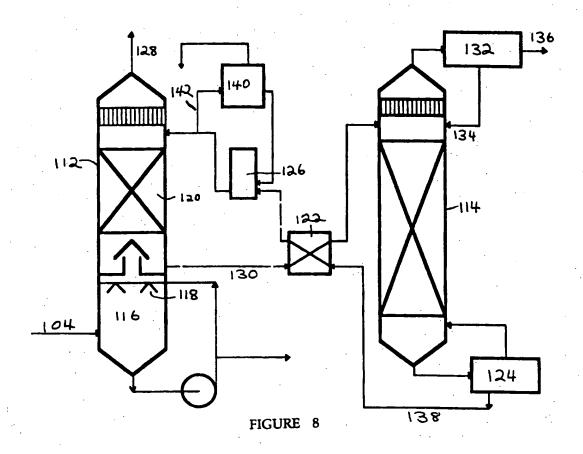


FIGURE 7



# THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

- 5 1. A process for the production of sulphuric acid comprising the steps of:
  - (a) providing a sulphuric acid precursor stream comprising oxygen and sulphur dioxide;
  - (b) feeding the sulphuric acid precursor stream to a sulphuric acid converter to produce a sulphuric acid stream and a gaseous stream containing unreacted sulphur dioxide; and,
  - (c) subjecting the gaseous stream to a regenerable sulphur dioxide recovery process to obtain a sulphur dioxide rich stream and a sulphur dioxide lean stream.

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- 2. The process as claimed in claim 1 wherein at least a portion of the sulphur dioxide rich stream is recycled and fed to the sulphuric acid converter.
- 20 3. The process as claimed in claim 1 wherein all of the sulphur dioxide rich stream is recycled and fed to the sulphuric acid converter.
  - 4. The process as claimed in claim 1 wherein the sulphuric acid converter comprises a sulphur dioxide converter having a single catalyst bed and the sulphur dioxide of the precursor stream is recycled through the process essentially to extinction.
  - 5. The process as claimed in claim 1 wherein the sulphuric acid converter comprises a sulphur dioxide converter having a single catalyst bed and a sulphur trioxide contactor, the sulphur dioxide converter produces a product stream containing sulphur trioxide, the

product stream is fed to the sulphur trioxide contactor to produce the sulphuric acid stream and the gaseous stream, and the sulphur dioxide lean stream contains less than 300 ppm sulphur dioxide.

- 5 6. The process as claimed in claim 1 wherein the sulphuric acid converter comprises a sulphur dioxide converter having two catalyst beds and the sulphur dioxide of the precursor stream is recycled through the process essentially to extinction.
- 7. The process as claimed in claim 1 wherein the sulphuric acid converter comprises a sulphur dioxide converter having up to two catalyst beds and a sulphur trioxide contactor, the sulphur dioxide converter produces a product stream containing sulphur trioxide, the product stream is fed to the sulphur trioxide contactor to produce the sulphuric acid stream and the gaseous stream, and the sulphur dioxide lean stream contains less than 300 ppm sulphur dioxide.
  - 8. A process for the production of sulphuric acid comprising the steps of:
- 20 (a) providing a first feed stream comprising sulphur dioxide;
  - (b) subjecting at least a portion of the first feed stream to a regenerable sulphur dioxide recovery process to obtain a first sulphur dioxide rich stream and a first sulphur dioxide lean stream;
  - (c) feeding at least a portion of the first sulphur dioxide rich stream and a second feed stream comprising oxygen to a sulphur dioxide converter to produce a product stream containing sulphur trioxide; and,

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(d) contacting the product stream with an aqueous solution to producea sulphuric acid stream and a gaseous stream containing unreacted sulphur dioxide and inert gases.

- 9. The process as claimed in claim 8 wherein the gaseous stream is subjected to the regenerable sulphur dioxide recovery process to obtain a second sulphur dioxide rich stream and a second sulphur dioxide lean stream.
- 10. The process as claimed in claim 9 wherein at least a portion of the second sulphur dioxide rich stream is recycled and fed to the sulphur dioxide converter as part of the first sulphur dioxide rich stream.

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- 11. The process as claimed in claim 8 wherein the gaseous stream is subjected to the regenerable sulphur dioxide recovery process to recover the sulphur dioxide from the gaseous stream and the sulphur dioxide recovered from the gaseous stream is recycled and fed to the sulphur dioxide converter as part of the first sulphur dioxide rich stream.
- 12. The process as claimed in claim 8 wherein all of the first feed stream is subjected to the regenerable sulphur dioxide recovery process.
- 13. The process as claimed in claim 8 wherein the sulphur dioxide converter comprises a single catalyst bed and the sulphur dioxide of the first feed stream is recycled through the process to extinction.
- 14. The process as claimed in claim 10 wherein the sulphur dioxide converter comprises a single catalyst bed, the sulphur dioxide of the first feed stream is recycled through the process and the sulphur dioxide lean streams contain less than 300 ppm sulphur dioxide.

15. The process as claimed in claim 8 wherein the sulphur dioxide converter comprises two catalyst beds and the sulphur dioxide of the first feed stream is recycled through the process to extinction.

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16. The process as claimed in claim 10 wherein the sulphur dioxide converter comprises two catalyst beds, the sulphur dioxide of the first feed stream is recycled through the process and the sulphur dioxide lean streams contain less than 300 ppm sulphur dioxide.

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- 17. A method of operating a sulphuric acid plant comprising:
  - (a) providing a feed stream comprising sulphur dioxide;
- (b) monitoring the sulphur dioxide concentration of the feed stream;

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(c) if the sulphur dioxide concentration of the feed stream is below the level required to achieve autothermal operation of a sulphur dioxide converter, subjecting at least a portion of the feed stream to a regenerable sulphur dioxide recovery process to obtain a first sulphur dioxide rich stream having a sulphur dioxide concentration sufficient to permit autothermal operation of the sulphur dioxide converter and a first sulphur dioxide lean stream;

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(d) feeding a stream selected from the group consisting of at least a portion of the feed stream, at least a portion of the first sulphur dioxide rich stream and mixtures thereof to the sulphur dioxide converter to produce a product stream containing sulphur trioxide and sulphur dioxide; and,

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(e) contacting the product stream with an aqueous solution to produce a sulphuric acid stream and a gaseous stream containing unreacted sulphur dioxide.

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18. The method as claimed in claim 17 wherein the gaseous

stream is subjected to the regenerable sulphur dioxide recovery process to obtain a second sulphur dioxide rich stream and a second sulphur dioxide lean stream.

- 5 19. The method as claimed in claim 18 wherein at least a portion of the second sulphur dioxide rich stream is recycled and fed to the sulphur dioxide converter as part of the first sulphur dioxide rich stream.
- 10 20. The method as claimed in claim 18 wherein all of the second sulphur dioxide rich stream is recycled and fed to the sulphur dioxide converter as part of the first sulphur dioxide rich stream.
- 21. The method as claimed in claim 17 wherein the sulphur dioxide converter comprises a single catalyst bed and the sulphur dioxide of the feed stream is recycled through the process essentially to extinction.
- 22. The method as claimed in claim 19 wherein the sulphur dioxide converter comprises a single catalyst bed, the sulphur dioxide of the feed stream is recycled through the process and the sulphur dioxide lean streams contain less than 300 ppm sulphur dioxide.
- 23. The method as claimed in claim 17 wherein the sulphur dioxide converter comprises two catalyst beds and the sulphur dioxide of the feed stream is recycled through the process essentially to extinction.
- 24. The method as claimed in claim 19 wherein the sulphur dioxide converter comprises two catalyst beds, the sulphur dioxide of the feed stream is recycled through the process and the sulphur

dioxide lean streams contain less than 300 ppm sulphur dioxide.

#### **ABSTRACT OF THE DISCLOSURE**

A process for the production of sulphuric acid comprises

providing a sulphuric acid precursor stream comprising oxygen and sulphur dioxide; feeding the sulphuric acid precursor stream to a sulphuric acid converter to produce a sulphuric acid stream and a gaseous stream containing unreacted sulphur dioxide; and, subjecting the gaseous stream to a regenerable sulphur dioxide recovery process to obtain a sulphur dioxide rich stream and a sulphur dioxide lean stream.